

Non-linear Elastic Response in Solid Helium: critical velocity or strain?

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Torsional oscillator experiments show evidence of mass decoupling in solid ^4He . This decoupling is amplitude dependent, suggesting a critical velocity for supersolidity. We observe similar behavior in the elastic shear modulus. By measuring the shear modulus over a wide frequency range, we can distinguish between an amplitude dependence which depends on velocity and one which depends on some other parameter like displacement. In contrast to the torsional oscillator behavior, the modulus depends on the magnitude of stress, not velocity. We interpret our results in terms of the motion of dislocations which are weakly pinned by ^3He impurities but which break away when large stresses are applied.

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Helium is a uniquely quantum solid and recent torsional oscillator (TO) measurements[1–8] provide evidence for a supersolid phase in hcp ^4He . At temperatures below about 200 mK the TO frequency increases, suggesting that some of the ^4He decouples from the oscillator. This evidence of non-classical rotational inertia (NCRI) inspired searches for other unusual thermal or mechanical behavior in solid helium. Heat capacity measurements[9, 10] do show a small peak near the onset temperature of decoupling, supporting the idea of a phase transition in solid ^4He . Mass flow is an obvious possible signature of supersolidity but experiments in this temperature range[11–13] show no pressure-induced flow through the solid (although recent measurements[14, 15] at higher temperatures showed intriguing behavior).

We recently made low frequency measurements[16, 17] of the shear modulus of hcp ^4He and found a large stiffening, with the same temperature dependence as the frequency changes seen in TO experiments. This modulus increase also had the same dependence on measurement amplitude and on ^3He concentration and, like the TO decoupling, its onset was accompanied by a dissipation peak. It is clear that the shear stiffening and the TO decoupling are closely related. Subsequent experiments[18] with ^3He showed similar elastic stiffening in the hcp phase below 0.4 K, but not in the bcc phase (the bcc phase exists only over a temperature range in ^4He). TO measurements with hcp ^3He did not, however, show any sign of a transition in the temperature range where the stiffening occurred, nor was a transition seen with bcc ^3He . The stiffening appears to depend on crystal structure (appearing in the hcp but not the bcc phase) while the TO frequency and dissipation changes occur only for the bcc solid, ^4He .

Although it is clear that solid ^4He shows unusual behavior below 200 mK, the interpretation in terms of supersolidity rests almost entirely upon torsional oscillator experiments. In addition to frequency changes which imply mass decoupling, two other features of the TO experiments are invoked as evidence of superflow. One is

the blocked annulus experiment[1, 19] in which NCRI is greatly reduced by inserting a barrier into the flow path, thus indicating that long-range coherent flow is involved. The other is the reduction of the NCRI fraction when the oscillation amplitude exceeds a critical value[1]. In analogy to superfluidity in liquid helium, this is interpreted in terms of a critical velocity v_c (of order $10 \mu\text{m/s}$), above which flow becomes dissipative. However, torsional oscillators are resonant devices and measurements made at a single frequency cannot distinguish an amplitude dependence which sets in at a critical velocity from one which begins at a critical displacement or a critical acceleration. A recent experiment[6] used a compound torsional oscillator which operated in two modes, allowing measurements to be made on the same solid ^4He sample at two different frequencies (496 and 1173 Hz). The amplitude dependence scaled somewhat better with velocity than with acceleration or displacement, supporting the superflow interpretation of TO experiments. However, recent measurements[20] in which one mode was driven at large amplitude while monitoring the low-amplitude response of the other mode gave unexpected results. The suppression of NCRI, as seen in the low amplitude mode, appeared to depend on the acceleration generated by the high amplitude mode, rather than on the velocity. To settle the question of whether the amplitude dependence seen in torsional oscillators reflects a critical velocity for superflow, measurements over a wider frequency range are needed.

We have made direct measurements[16] of the amplitude dependence of the shear modulus, μ , of hcp ^4He in a narrow gap of thickness D . An AC voltage, V , with angular frequency $\omega = 2\pi f$, is applied to a shear transducer (with piezoelectric coefficient d_{15}) to generate a displacement $\delta x = d_{15}V$ at its surface. This produces a quasi-static shear strain in the helium, $\epsilon = \delta x/D$. The resulting stress, σ , generates a charge, q , and thus a current, $I = \omega q$, in a second transducer on the opposite side of the gap. The shear modulus $\mu = \sigma/\epsilon$ is then proportional to I/fV . In contrast to torsional oscillator

measurements, this technique is non-resonant and so we could measure the shear modulus over a wide frequency range, from a few Hz (a limit set by preamplifier noise) to a maximum of 2500 Hz (due to interference from the first acoustic resonance in our cell, around 8 kHz). The technique is very sensitive, allowing us to make modulus measurements at strains as low as $\epsilon \approx 10^{-9}$, corresponding to stresses $\sigma \approx 0.02$ Pa. The drive voltage can be increased substantially without heating the sample, so measurements can be made at strains up to $\epsilon \approx 10^{-5}$, allowing us to study the amplitude dependence, including hysteretic effects, at low temperatures.

Figure 1 shows the temperature dependence of the normalized shear modulus μ/μ_0 for an hcp ^4He sample at a pressure of 38 bar and a frequency of 2000 Hz. The crystal was grown from standard isotopic purity ^4He (containing about 0.3 ppm ^3He) using the blocked capillary method. A piezoelectric shear stack[21] was used to generate strains in a narrow gap ($D = 500 \mu\text{m}$) between it and a detecting transducer. Other experimental details are the same as in refs. 16 to 18. The curves in Fig. 1 correspond to different transducer drive voltages, i.e. to different strains, and were measured during cooling from a temperature of 0.7 K. The shear modulus increases at low temperature, with $\Delta\mu/\mu_0 \approx 17\%$ at the lowest amplitude. Similar stiffening was seen in all hcp ^4He crystals[16, 17], although the onset temperature was lower in crystals of higher isotopic purity and the magnitude of the modulus change varied by a factor of about 2 from sample to sample (the TO NCRI varies much more, by a factor of 1000, although its temperature dependence is always essentially the same[18]). The amplitude dependence of the shear modulus stiffening is essentially the same as that of the TO NCRI. At the lowest drive voltages and temperatures, both are independent of amplitude but they decrease at high amplitudes. The onset of stiffening shifts to lower temperatures at high amplitudes, as does the onset of TO decoupling. However, in the case of elastic measurements, it is more natural to think of this behavior in terms of a critical stress (proportional to the strain, i.e. to transducer displacement), rather than a critical velocity.

The amplitude dependence of the shear modulus is shown in more detail in Fig. 2. Open circles show the modulus at 48 mK, taken from the temperature sweeps (i.e. the points marked by open circles in Fig. 1). The solid circles are the modulus measured when the drive voltage was reduced at fixed temperature (48 mK), after cooling from high temperature at the highest drive amplitude ($3 V_{pp}$). Figure 2 also shows the corresponding amplitude dependence at temperatures well above the shear modulus anomaly (open squares are data at 700 mK from the temperature sweeps of Fig. 1; solid squares are from amplitude sweeps at 800 mK). The data taken with the two protocols agree very well. The critical drive voltage (where the modulus becomes amplitude depen-

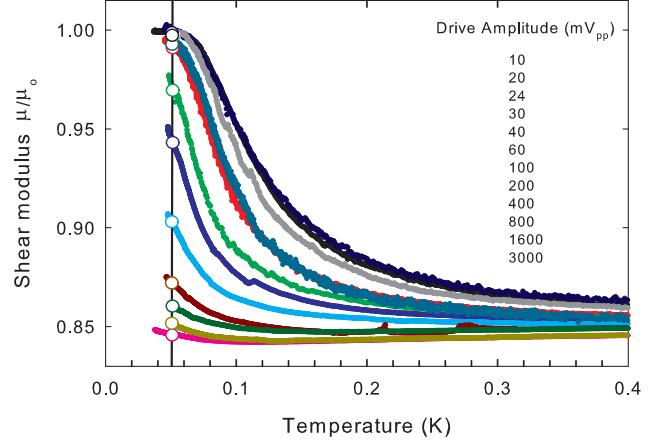


FIG. 1: Temperature and amplitude dependence of the shear modulus in a solid ^4He sample at 38 bar, grown by the blocked capillary method. The modulus was measured at 2000 Hz for transducer drive voltages (peak to peak) from 10 mV to 3 V. Values are normalized by μ_0 the low temperature value at the lowest drive voltage.

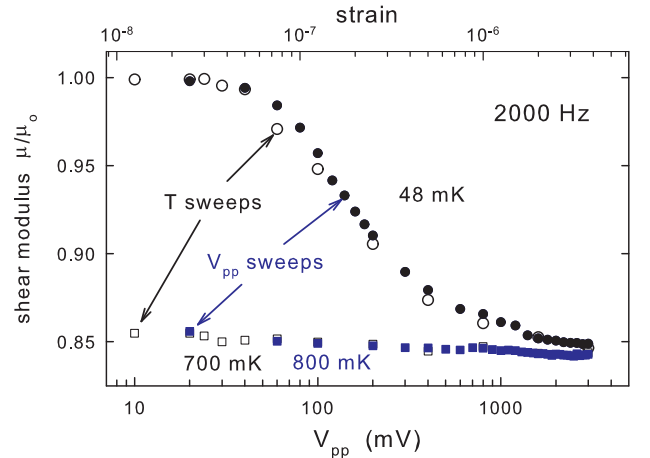


FIG. 2: Low and high temperature shear modulus for the crystal of Fig. 1, as a function of drive voltage (bottom axis) or strain (top axis). The open symbols are taken from temperature sweeps at different drive levels (the data of Fig. 1). The solid symbols are taken while decreasing the drive voltage at fixed temperature.

dent) is around 30 mV_{pp} (corresponding to $\sigma \approx 4 \times 10^{-8}$, $\sigma \approx 0.8$ Pa) and at the highest drive levels ($3 V_{pp}$ corresponding to $\sigma \approx 80$ Pa) the stiffening is almost completely suppressed. The amplitude dependence of the modulus closely resembles that of the NCRI in TO experiments, behavior which is attributed to a superflow critical velocity (typically around $10 \mu\text{m/s}$). However, even in TO measurements there are inertial stresses in the helium, produced by its acceleration, which could lead to the observed amplitude dependence.

A number of experiments[6, 19, 22, 23] have shown that

the TO amplitude dependence is hysteretic at low temperatures. If a sample is cooled at high oscillation amplitude, the apparent NCRI is small. When the amplitude is reduced at low temperature, the TO frequency (NCRI) rises, becoming constant below some critical amplitude. When the drive is then increased at low temperature, the NCRI does not begin to decrease at the critical amplitude - it remains essentially constant at substantially larger drives. This hysteresis between data taken while decreasing and increasing the drive amplitude disappears at temperatures above about 70 mK.

Figure 3 shows the corresponding hysteresis in the shear modulus. At 120 mK the maximum stiffening is about half as large as at 36 mK and already depends on amplitude at the lowest strains shown. The modulus measured when the amplitude is reduced (open circles) and when it is subsequently increased (solid circles) agree. Hysteresis appears when the sample is cooled below 60 mK and is nearly temperature independent below 45 mK. Figure 3 shows this hysteresis at 36 mK. The sample was cooled from high temperature while driving at high amplitude ($3 V_{pp}$). The amplitude was then lowered at 36 mK (open circles) which resulted in a shear modulus increase $\Delta\mu/\mu_o$ of about 15%. When the amplitude was then raised (solid circles), the modulus remained constant to much higher amplitude, the same behavior seen for the NCRI in TO experiments. At very high amplitudes (above about $1 V_{pp}$, corresponding to $\epsilon \approx 10^{-6}$, $\sigma \approx 20$ Pa) the modulus decreased, nearly closing the hysteresis loop. After each change we waited 2 minutes for the modulus to stabilize at the new amplitude. The only region where we observed further time dependence was while increasing the amplitude at drive levels above $1 V_{pp}$. The modulus decrease was sharper when we waited longer at each point. In acoustic resonance measurements[17], we found that even larger stresses ($\sigma \approx 700$ Pa) produced irreversible changes which only disappeared after annealing above 0.5 K.

The only obvious mechanism which can produce shear modulus changes as large as those shown in Figs. 1 to 3 involves the motion of dislocations[24]. At low temperatures, dislocations are pinned by ^3He impurities and the intrinsic modulus is measured[17]. As the temperature is raised, ^3He impurities thermally unbind from the dislocations, allowing them to move and reducing the modulus. At high amplitudes, elastic stresses can also tear the dislocations away from the impurities. The hysteresis seen in Fig. 3 can be understood if, when a crystal is cooled at high strain amplitudes, the rapid motion of dislocations prevents ^3He atoms from attaching to them. When the drive is reduced at low temperatures, impurities can bind, thus pinning the dislocations and increasing the modulus. Once the ^3He impurities bind, the pinning length of dislocations is smaller and larger stresses are required to unpin them so the modulus retains its intrinsic value to much higher amplitudes. The critical amplitude for

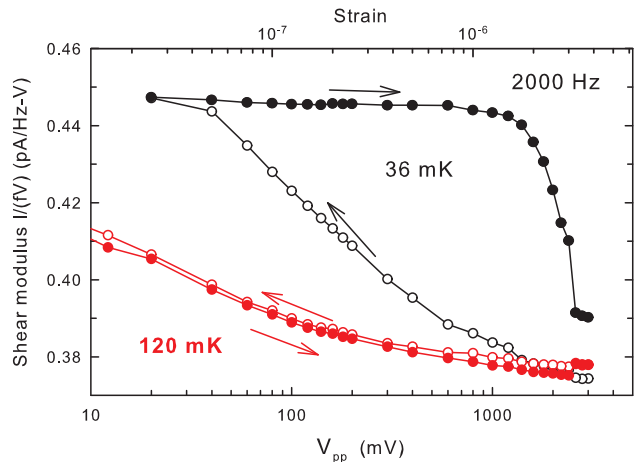


FIG. 3: Hysteresis between the shear modulus measured while decreasing and while increasing the drive amplitude at low temperature (36 mK). At 120 mK the amplitude dependence is reversible.

this stress-induced breakaway can be estimated[25, 26] if the dislocation length and impurity binding energy are known. In single crystals of helium, a typical dislocation network pinning length[27] is ($L \sim 5 \mu\text{m}$), and dislocations would break away from a ^3He impurity at strain $\epsilon \approx 3 \times 10^{-7}$. Our crystals are expected to have higher dislocation densities and smaller network lengths, so breakaway would occur at higher strains as the amplitude is increased. Measurements with different ^3He concentrations would be useful to confirm that the amplitude dependence is due to this mechanism.

This interpretation of the shear modulus behavior involves elastic stress (which is proportional to strain) rather than velocity, and so is at odds with the interpretation of the TO amplitude dependence in terms of a superfluid-like critical velocity. Since we can make modulus measurements over a wide frequency range, we can unambiguously distinguish between an amplitude dependence which scales with stress (or strain) and one which depends on velocity. Figure 4 shows the modulus at 18 mK, measured at three different frequencies (2000, 200 and 20 Hz) as the drive amplitude was reduced from its maximum value. In Fig. 4a the modulus is plotted vs. shear strain ϵ (calibrated using the low temperature piezoelectric coefficient of the shear stack, $d_{15} = 1.25 \text{ nm/V}$) and in Fig. 4b the same data is plotted versus the corresponding velocities ($v = \omega \epsilon D$). The amplitude dependence scales much better with strain than with velocity (and the scaling with acceleration is even less satisfactory). The critical strain appears to be slightly larger at lower frequency.

It is clear that the amplitude dependence of the shear modulus is most closely associated with stress (or strain) amplitude, rather than with a superfluid-like critical ve-

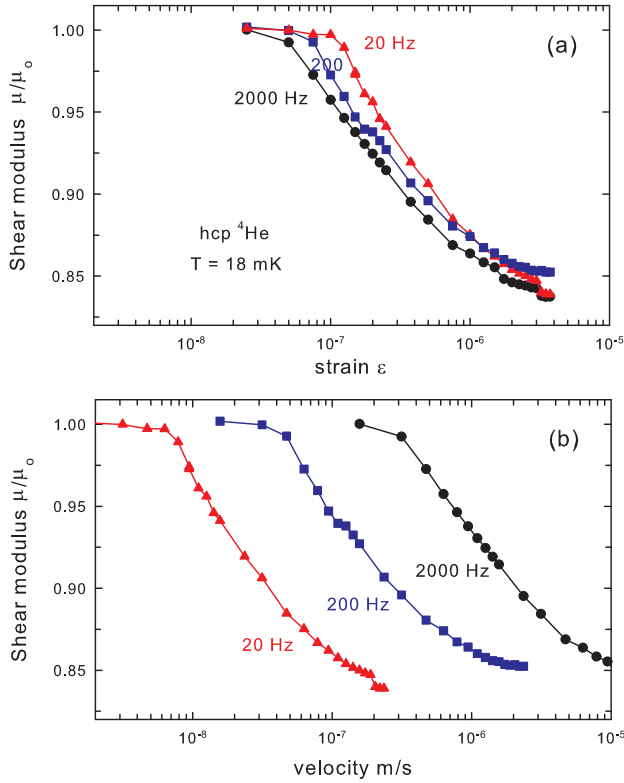


FIG. 4: Scaling of the shear modulus amplitude dependence with (a) strain and (b) velocity for three different frequencies: 20 Hz (triangles), 200 Hz (squares) and 2000 Hz (circles).

locity. The many similarities to the TO behavior (e.g. the dependence on temperature, ^3He concentration and frequency, the amplitude dependence and its hysteresis) suggest that the apparent velocity dependence of the NCRI may have a similar origin, e.g. in inertial stresses which exceed the critical value for the shear modulus. However, estimates of the inertial stress corresponding to TO critical velocities give values significantly lower than the critical stress for the shear modulus. For an annular TO geometry, the maximum inertial stress can be estimated as $\sigma = \rho t \omega v / 2$, where ρ is the helium density, t is the width of the annular channel, ω is the angular frequency of the TO and v is the oscillation velocity. For the oscillator of ref. 1, we estimate $\sigma \approx 0.002$ to 0.015 Pa at the apparent critical velocities of 5 to $38 \mu\text{m/s}$. Measurements in an open cylindrical TO[23] show amplitude dependence at velocities corresponding to inertial stresses below 0.01 Pa, also much smaller than the stresses at which we observe amplitude dependence in the shear modulus. There is also other evidence that the TO frequency changes and dissipation are not just mechanical consequences of the modulus changes. First, the apparent NCRI is too large to be explained simply by mechanical stiffening of the torsional oscillator[18, 23]. Secondly, comparable modulus changes in hcp ^3He are

not reflected in the corresponding TO behavior[18]. The existence of a critical velocity for superflow in solid helium can only be definitively shown if TO measurements can be made over a wide range of frequency and/or TO geometries.

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